Diagrammatic semiclassical laser theory

Oleg Zaitsev[1,2,*] and Lev Deych[3,†]
[1] *Physikalisches Institut der Universität Bonn, Nußallee 12, 53115 Bonn, Germany*
[2] *Fachbereich Physik, Universität Duisburg-Essen, Lotharstraße 1, 47048 Duisburg, Germany*
[3] *Physics Department, Queens College of City University of New York, Flushing, New York 11367, USA*
(Received 9 December 2009; published 22 February 2010)

We derive semiclassical laser equations valid in all orders of nonlinearity. With the help of a diagrammatic representation, the perturbation series in powers of electric field can be resummed in terms of a certain class of diagrams. The resummation makes it possible to take into account a weak effect of population pulsations in a controlled way while treating the nonlinearity exactly. The proposed laser theory reproduces the all-order nonlinear equations in the approximation of constant population inversion and the third-order equations with population-pulsation terms as special cases. The theory can be applied to arbitrarily open and irregular lasers, such as random lasers.

DOI: 10.1103/PhysRevA.81.023822 PACS number(s): 42.55.Ah, 42.55.Zz

I. INTRODUCTION

Interest in random lasers with coherent feedback [1,2] and lasers based on chaotic microresonators without mirrors [3] revealed a number of shortcomings of conventional laser theory [4,5] that complicated its application to such systems. Among the properties that characterize random and chaotic lasers are strong openness, irregular spatial dependence of the refractive index, and possibly a nontrivial shape of the resonator. Lasing in these systems can be accompanied by strong coupling between the modes, which requires a more careful treatment of nonlinear effects than is necessary for regular lasers.

An essential part of a laser description is the choice of an appropriate basis of normal modes in which the electromagnetic field and other system functions can be expanded. In an open system, it is not possible to define a Hermitian eigenvalue problem whose eigenfunctions would form an orthogonal basis. Instead, one has to introduce a biorthogonal system of so-called quasimodes as left and right eigenfunctions of a non-Hermitian operator. A number of methods to construct quasimodes have been discussed in the literature. Among the earliest are the Fox-Li modes [6–8], which are useful in resonators with a preferred propagation direction and clearly defined transverse plane. In a more general setting, there have been attempts to use solutions of the wave equation that satisfy outgoing boundary conditions at infinity (Siegert-Gamow boundary conditions) with complex eigenfrequencies corresponding to scattering resonances [9–11]. However, these modes diverge at infinity, which makes it problematic to use them as a basis [9], but some ways around this problem have been discussed in Refs. [10] and [11]. Another possibility is to use the so-called system-and-bath-type approaches [12,13], where cavities are described by an orthogonal system of wave functions of a Hermitian problem with an independent sets of modes introduced for outside of the resonator. The openness of the cavity in this approach is reproduced by introducing coupling between a discrete set of inside modes and the continuous spectrum of outside modes. This way, the modal expansion of the field becomes possible both inside and outside of the cavity.

In the present work, we use a different approach based on keeping the spectral parameter of the outside outgoing field real while making the inside field satisfy continuity conditions at the boundary of the cavity. This approach also results in the discrete spectrum of the cavity field with complex-valued frequencies, but because the outside field is forced to depend on the real spectral parameter, it does not diverge and is characterized by a constant flux. This approach was used in Ref. [14] for a special kind of vibration problems and was adapted for lasers in Ref. [15], where these modes were dubbed the constant-flux (CF) modes. One can introduce two adjoint biorthogonal systems of CF modes, which can be used to represent a field inside the cavity.

In the standard semiclassical laser theory [4,5], lasing modes are usually taken to coincide with quasimodes of the respective cavities, and their amplitudes and frequencies are found from equations based on perturbation expansions containing terms linear and cubic in the field. In random lasers, this picture needs to be revised. First, it has been shown [16,17] that normal modes in the presence of gain differ from the passive modes even in the linear approximation if the refractive index and/or unsaturated population inversion are nonuniform. Second, it has been realized [18–20] that self- and cross-saturation coefficients before the cubic terms can have different statistical properties in different systems, leading to different mode statistics. Finally, it has been pointed out [15,21] that nonlinear effects can significantly contribute to modification of the lasing modes compared to those of the empty cavity. A theory suggested in Refs. [15,21] allowed for self-consistent calculations of not only lasing frequencies but also of the spatial distributions of the respective modes. By neglecting pulsations of the population inversion, the authors of Refs. [15,21] were able to derive equations for field amplitudes and frequencies beyond the usual third-order approximation.

In the present work, we also generalize the conventional laser theory, but, unlike the approach of Refs. [15,21], we do not begin by introducing a special approximation for population inversion. Instead, we carry out the perturbation

*oleg.zaitsev@uni-bonn.de
†lev.deych@qc.cuny.edu

expansion up to the infinite order in the field, keeping all the terms that do not have fast temporal oscillations. This also includes a part of population-pulsation contributions that is consistent with the slowly-varying-envelope approximation. The classification of the resulting terms becomes possible as a result of a special diagram technique developed to represent the terms of the expansion. However, this technique differs significantly from the usual Feynman diagrams widely used in solid-state and high-energy physics, because we have to deal with terms of ever-increasing degrees of nonlinearity. Thus, diagrams in our technique are not used to literally represent each term of the expansion, but play a more limited role as a tool assisting in the classification of the terms. Nevertheless, this technique allows for standard division of diagrams into connected and disconnected ones, with disconnected diagrams submitting to easy resummation in terms of only connected ones. The connected diagrams can be classified according to the order of magnitude of the population pulsations. The resulting laser equations generalize the nonlinear equations of Refs. [15,21] in two respects. First, equations derived in this article are dynamic, allowing for the study of time dependence of the amplitudes, whereas the equations of Refs. [15,21] can describe only stationary lasing output. Second, our equations incorporate terms responsible for the population-pulsation contribution in all orders of the perturbation theory; the equations of Refs. [15,21] are reproduced in our approach if only the lowest-order diagram is taken into account.

The structure of our article is as follows. In Sec. II, we recall the definition and properties of the constant-flux quasimodes of open system. Standard semiclassical laser equations are written in Sec. III in frequency representation for later convenience. Coupled equations for electric field, polarization, and population inversion are reduced to equations for the field alone in Sec. IV using infinite-order perturbation theory. In Sec. V, we formulate the diagrammatic technique and resum the perturbation expansion in terms of connected diagrams. In Sec. VI, we reproduce the results of linear theory, third-order theory with population-pulsation terms, and all-order nonlinear theory in the constant-inversion approximation. Finally, we write corrections to the all-order theory that are of the first order in the population pulsations.

II. CONSTANT-FLUX QUASIMODES

We consider an open system defined by real dielectric constant $\epsilon(\mathbf{r})$, with $\epsilon = 1$ outside of the system's boundary. In the Coulomb gauge $\nabla \cdot [\epsilon(\mathbf{r})\mathbf{E}(\mathbf{r}, t)] = 0$, an electric field $\mathbf{E}(\mathbf{r}, t)$ is governed by the wave equation

$$\epsilon(\mathbf{r})\frac{\partial^2}{\partial t^2}\mathbf{E} + \nabla \times (\nabla \times \mathbf{E}) = 0, \tag{1}$$

where Gaussian units with the velocity of light in vacuum $c = 1$ are used.

In the absence of gain and absorption, the field will decay in time as a result of the openness. It is convenient to represent the decaying field as a superposition of certain normal modes, the quasimodes, that have only outgoing components outside the system. These modes can be constructed as families of CF states [15] $\psi_k(\mathbf{r}, \omega)$ depending on a real continuous parameter ω. Explicitly, the CF modes satisfy the differential equation

$$\nabla \times [\nabla \times \psi_k(\omega)] = \omega^2 \psi_k(\omega) \tag{2}$$

in the exterior of the cavity with the outgoing-wave boundary conditions at infinity. Inside the system, the same state satisfies a different equation:

$$\frac{1}{\sqrt{\epsilon(\mathbf{r})}}\nabla \times \left[\nabla \times \frac{\psi_k(\omega)}{\sqrt{\epsilon(\mathbf{r})}}\right] = \Omega_k^2(\omega)\psi_k(\omega). \tag{3}$$

For each ω, the complex eigenfrequency $\Omega_k(\omega)$ is quantized, as the eigenfunctions are required to match smoothly at the interface.

Conjugate wave functions $\phi_k(\mathbf{r}, \omega)$ obey Eq. (2) outside with the incoming-wave boundary conditions and the equation

$$\frac{1}{\sqrt{\epsilon(\mathbf{r})}}\nabla \times \left[\nabla \times \frac{\phi_k(\omega)}{\sqrt{\epsilon(\mathbf{r})}}\right] = \left[\Omega_k^2(\omega)\right]^* \phi_k(\omega) \tag{4}$$

inside the system. The CF functions and their conjugates are biorthogonal and can be chosen to satisfy the condition

$$\int_{\mathcal{I}} d\mathbf{r}\, \phi_k^*(\mathbf{r}, \omega) \cdot \psi_{k'}(\mathbf{r}, \omega) = \delta_{kk'}, \tag{5}$$

where the integration is over the interior $\mathcal{I}$.

A Fourier component $\mathbf{E}_\omega(\mathbf{r})$ of the internal field can be expanded in the CF modes as

$$\mathbf{E}_\omega(\mathbf{r}) = \epsilon^{-1/2}(\mathbf{r}) \sum_k a_k(\omega)\psi_k(\mathbf{r}, \omega), \tag{6}$$

$$a_k(\omega) = \int_{\mathcal{I}} d\mathbf{r} \sqrt{\epsilon(\mathbf{r})}\phi_k^*(\mathbf{r}, \omega) \cdot \mathbf{E}_\omega(\mathbf{r}). \tag{7}$$

When continued to the exterior, this expansion yields a wave at the frequency ω propagating in the free space away from the system. In a stationary lasing regime, ω becomes subjected to an equation, which has a discrete set of solutions corresponding to the frequencies of lasing modes.

III. SEMICLASSICAL LASER EQUATIONS

In the semiclassical theory of lasers [4,5], the fields are described classically at the level of Maxwell equations, and the active medium is treated by quantum mechanics. To this end, the wave Eq. (1) is written with a source term, the polarization $\mathbf{P}(\mathbf{r}, t)$ of the gain medium:

$$\epsilon(\mathbf{r})\frac{\partial^2}{\partial t^2}\mathbf{E} + \nabla \times (\nabla \times \mathbf{E}) = -4\pi \frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{r}, t). \tag{8}$$

In the simplest model, the active medium is a collection of homogeneously broadened two-level atoms. Their state is fully described by $\mathbf{P}(\mathbf{r}, t)$ and the population-inversion density $\Delta n(\mathbf{r}, t)$ (difference between populations of the upper and lower levels per unit volume). These functions satisfy the equations of motion [5]

$$\left(\frac{\partial^2}{\partial t^2} + 2\gamma_\perp \frac{\partial}{\partial t} + \nu^2\right)\mathbf{P} = -2\nu \frac{d^2}{\hbar}\mathbf{E}(\mathbf{r}, t)\Delta n(\mathbf{r}, t), \tag{9}$$

$$\frac{\partial}{\partial t}\Delta n - \gamma_\parallel[\Delta n_0(\mathbf{r}, t) - \Delta n] = \frac{2}{\hbar\nu}\mathbf{E}(\mathbf{r}, t) \cdot \frac{\partial}{\partial t}\mathbf{P}(\mathbf{r}, t), \tag{10}$$

where d is the magnitude of the atomic dipole matrix element, ν is the atomic transition frequency, and $\gamma_\perp$ ($\gamma_\parallel$) is the polarization (population-inversion) decay rate. If the right-hand side of Eq. (10) vanishes, Δn relaxes to the unsaturated population inversion $\Delta n_0(\mathbf{r}, t)$, which is a measure of the pump strength. The coupled Eqs. (8)–(10) determine, in principle, electric field in the system, if $\Delta n_0(\mathbf{r}, t)$ is given.

It is convenient, at this stage, to rewrite the equations of motion in the frequency representation. The Fourier-transformed variables are given in the form

$$\mathbf{E}(\mathbf{r}, t) = \frac{1}{\pi}\mathrm{Re}\int_0^\infty d\omega\, \mathbf{E}_\omega(\mathbf{r})e^{-i\omega t}, \tag{11}$$

$$\mathbf{P}(\mathbf{r}, t) = \frac{1}{\pi}\mathrm{Re}\int_0^\infty d\omega\, \mathbf{P}_\omega(\mathbf{r})e^{-i\omega t}, \tag{12}$$

$$\Delta n(\mathbf{r}, t) = \frac{1}{2\pi}\int_{-\infty}^\infty d\omega\, \Delta n_\omega(\mathbf{r})e^{-i\omega t}, \tag{13}$$

which facilitates application of the rotating-wave approximation. Additionally, we assume that $\mathbf{E}_\omega$ and $\mathbf{P}_\omega$ are negligible outside of a small vicinity of ν. Then the frequency squared can be approximated as $\omega^2 = (\nu + \omega - \nu)^2 \approx \nu^2 + 2\nu(\omega - \nu)$, and the lasing equations become effectively the first-order differential equations in time,

$$-\epsilon(\mathbf{r})(-\nu^2 + 2\nu\omega)\mathbf{E}_\omega + \nabla \times (\nabla \times \mathbf{E}_\omega) = 4\pi\nu^2\mathbf{P}_\omega, \tag{14}$$

$$[-i(\omega - \nu) + \gamma_\perp]\mathbf{P}_\omega = -i\frac{d^2}{2\pi\hbar}\int_0^\infty d\omega'\, \mathbf{E}_{\omega'}\Delta n_{\omega-\omega'}, \tag{15}$$

$$(-i\omega + \gamma_\parallel)\Delta n_\omega = 2\pi\gamma_\parallel\Delta n_0(\mathbf{r})\delta(\omega) - \frac{i}{\pi\hbar} \times \int_0^\infty d\omega'(\mathbf{E}^*_{\omega'-\omega}\cdot\mathbf{P}_{\omega'} - \mathbf{E}_{\omega'+\omega}\cdot\mathbf{P}^*_{\omega'}), \tag{16}$$

where the pump $\Delta n_0(\mathbf{r})$ is assumed constant in time.

IV. ALL-ORDER PERTURBATION THEORY

A. Expansions for polarization and population inversion

Equations (14)–(16) can be reduced to an equation for the electric field alone using perturbation theory in the field amplitude. In particular, one needs to construct an expansion of $\mathbf{P}_\omega(\mathbf{r})$ in the (odd) powers of the field using Eqs. (15) and (16). Then this expansion is substituted in Eq. (14), producing the required equation for the field. In the conventional laser theory [4,5], $\mathbf{P}_\omega(\mathbf{r})$ is expanded up to the third order in $\mathbf{E}_\omega(\mathbf{r})$, which yields the saturation terms in the rate equations. In this article, we carry out the expansion up to an arbitrary order in the field's amplitude and use diagrammatic method to sort out the respective terms.

We begin by neglecting the quadratic terms in Eq. (16), which gives the zero-order expression for the population inversion: $\Delta n^{(0)}_\omega = 2\pi\Delta n_0(\mathbf{r})\delta(\omega)$. By substituting this expression in the polarization Eq. (15), we obtain the first-order term for polarization: $\mathbf{P}^{(1)}_\omega = -i(d^2/\hbar\gamma_\perp)D(\omega)\Delta n_0(\mathbf{r})\mathbf{E}_\omega$, where

$$D(\omega) \equiv \left(1 - i\frac{\omega - \nu}{\gamma_\perp}\right)^{-1}. \tag{17}$$

This expression is substituted back in Eq. (16), from which the second-order correction to the inversion $\Delta n^{(2)}_\omega$ is determined. This iteration procedure yields the perturbation series for polarization and population inversion,

$$\mathbf{P}_\omega(\mathbf{r}) = \sum_{q\ \mathrm{odd}} \mathbf{P}^{(q)}_\omega(\mathbf{r}), \tag{18}$$

$$\Delta n_\omega(\mathbf{r}) = \sum_{q\ \mathrm{odd}} \Delta n^{(q-1)}_\omega(\mathbf{r}), \tag{19}$$

in odd and even powers of the electric field, respectively. Henceforth, we restrict the calculations to the case of scalar field. The general terms of these expansions are derived by induction in Appendix A. The resulting expressions are

$$\begin{aligned} P^{(q)}_\omega &= 2i\hbar\gamma_\parallel\frac{A^{(q+1)/2}}{\pi^{q-1}}\Delta n_0(\mathbf{r})D(\omega)\int d\omega_1\cdots d\omega_{q-1}E_{\omega_1}E^*_{\omega_2}\cdots E_{\omega_{q-2}}E^*_{\omega_{q-1}}E_{\omega-\omega_1+\omega_2-\cdots-\omega_{q-2}+\omega_{q-1}}D_\parallel(\omega_1-\omega_2)\\ &\quad\times D_\parallel(\omega_1-\omega_2+\omega_3-\omega_4)\cdots D_\parallel(\omega_1-\omega_2+\cdots+\omega_{q-2}-\omega_{q-1})[D(\omega_1)+D^*(\omega_2)][D(\omega_1-\omega_2+\omega_3)\\ &\quad+D^*(\omega_2-\omega_1+\omega_4)]\cdots[D(\omega_1-\omega_2+\omega_3-\cdots-\omega_{q-3}+\omega_{q-2})+D^*(\omega_2-\omega_1+\omega_4-\cdots-\omega_{q-4}+\omega_{q-1})], \end{aligned} \tag{20}$$

$$\begin{aligned} \Delta n^{(q+1)}_\omega &= 2\frac{A^{(q+1)/2}}{\pi^q}\Delta n_0(\mathbf{r})D_\parallel(\omega)\int d\omega_1\cdots d\omega_q E_{\omega_1}E^*_{\omega_2}\cdots E^*_{\omega_{q-1}}E_{\omega_q}E^*_{-\omega+\omega_1-\omega_2+\cdots-\omega_{q-1}+\omega_q}D_\parallel(\omega_1-\omega_2)\\ &\quad\times D_\parallel(\omega_1-\omega_2+\omega_3-\omega_4)\cdots D_\parallel(\omega_1-\omega_2+\cdots+\omega_{q-2}-\omega_{q-1})[D(\omega_1)+D^*(\omega_2)][D(\omega_1-\omega_2+\omega_3)\\ &\quad+D^*(\omega_2-\omega_1+\omega_4)]\cdots[D(\omega_1-\omega_2+\omega_3-\cdots-\omega_{q-3}+\omega_{q-2})+D^*(\omega_2-\omega_1+\omega_4-\cdots-\omega_{q-4}+\omega_{q-1})]\\ &\quad\times D(\omega_1-\omega_2+\omega_3-\cdots-\omega_{q-1}+\omega_q)+\mathrm{c.c.}(\omega\to-\omega), \end{aligned} \tag{21}$$

where q is odd. The notation c.c.$(\omega \to -\omega)$ stands for the first part of the equation to which complex conjugation accompanied by change of the sign of ω was applied. We also introduced the following definitions:

$$A \equiv -\frac{d^2}{2\hbar^2\gamma_\perp\gamma_\parallel}, \tag{22}$$

$$D_{\parallel}(\omega) \equiv \left(1 - i\frac{\omega}{\gamma_{\parallel}}\right)^{-1}. \qquad (23)$$

These expressions describe nonlinear ($q \geqslant 3$) corrections to polarization and population inversion, which are used to obtain equations for the field amplitudes.

B. Equations for electric field

Equation for the amplitudes (7) of normal modes,

$$[\Omega_k(\omega) - \omega]a_k(\omega) = 2\pi\nu \int_{\mathcal{I}} d\mathbf{r}\epsilon^{-1/2}(\mathbf{r})\phi_k^*(\mathbf{r},\omega)P_\omega(\mathbf{r}), \qquad (24)$$

follows from Eq. (14) and the biorthogonality condition (5). The right-hand side is a sum of the infinite number of nonlinear corrections to the polarization and is, therefore, a functional of *all* amplitudes $a_k(\omega)$. By definition, a lasing mode is such a solution that, in the limit $t \to \infty$, approaches a purely harmonic form, $\propto \exp(-i\omega_k t)$. Here ω_k is some frequency that is determined self-consistently from Eq. (24). In the frequency domain, these solutions are characterized by δ-functional frequency dependence $\propto \delta(\omega - \omega_k)$.

Generally speaking, lasing modes are different from the quasimodes of the passive system. Indeed, an attempt to search for the solutions of Eq. (24) in the form $a_k(\omega) = a_k\delta(\omega - \omega_k)$ leads, after the frequency integration (20), to the equation

$$\begin{aligned}&[\Omega_k(\omega_k) - \omega_k]a_k\delta(\omega - \omega_k)\\ &\quad = \sum_{q\text{ odd}}\sum_{k_1,\cdots,k_q} W^{(q)}_{k,k_1,\ldots,k_q} a_{k_1}a^*_{k_2}\ldots a^*_{k_{q-1}}a_{k_q}\\ &\qquad \times \delta\big(\omega - \omega_{k_1} + \omega_{k_2} - \cdots + \omega_{k_{q-1}} - \omega_{k_q}\big),\end{aligned} \qquad (25)$$

where the coefficients $W^{(q)}_{k,k_1,\ldots,k_q}$ are functions of the frequencies $\omega_k, \omega_{k_1}, \ldots, \omega_{k_q}$. Clearly, most of the terms on the right-hand side contain the δ functions that are different from $\delta(\omega - \omega_k)$ on the left-hand side. In the time domain, these terms would introduce oscillations with frequencies different from ω_k. This means that, in general, no stationary lasing solutions exist unless for some reason the terms oscillating at the "beat" frequencies are small and can be neglected. Usually the selection of the slowly changing contributions to Eq. (25) is done by leaving only terms with pairwise coinciding indices k_i, so that the respective frequency differences cancel out (more on this procedure can be found later). However, because the number of ω_{k_i} in this equation is odd, one will always remain with the expression $\delta(\omega - \omega_{k'})$, where k' is the index of one of the remaining uncanceled frequencies.

This problem can only be resolved by requiring that, in a given lasing mode l, each contribution a_{lk} of the quasimode k oscillates at the same frequency, and the general solution for a_k takes the form of

$$a_k(\omega) = \pi\sum_l a_{lk}\delta(\omega - \omega_l). \qquad (26)$$

Amplitudes a_{lk} can be shown to obey the equation

$$[\Omega_k(\omega_l) - \omega_l]a_{lk} = \sum_{k'} V^{(l)}_{kk'}a_{lk'}, \qquad (27)$$

where the coefficients $V^{(l)}_{kk'}$ depend on all frequencies and amplitudes. An explicit expression for $V^{(l)}_{kk'}$ is given later. One can see from this equation that the lasing modes are those combinations of the quasimodes that diagonalize the matrix $V^{(l)}_{kk'}$, whereas lasing frequencies are real eigenvalues of this matrix [16,17].

One has to realize, though, that Eq. (27) is not an ordinary eigenvalue problem, because the matrix $V^{(l)}_{kk'}$ itself depends on the amplitudes a_{lk}. Unlike linear eigenvalue problems, which determine frequencies for which nonzero solutions for the amplitudes can exist, by solving Eq. (27) one shall be able to find the frequencies as well as the field amplitudes. This is possible because the requirement that the respective eigenfrequencies must be real provides an additional constraint on solutions of Eq. (27) [21,22].

If matrix $V^{(l)}_{kk'}$ is calculated in the constant-inversion approximation, Eq. (27) reproduces the main result of Ref. [23]. However, while the derivation of this equation in Ref. [23] is only valid in the strictly stationary limit, the arguments presented here can be extended to a nonstationary case. Indeed, we can repeat these arguments for a weakly nonstationary situation, requiring that the amplitudes a_{lk} be slowly changing functions of time. Formally, we replace the mode expansion (26) with $a_k(\omega) = \sum_l a_{lk}(\omega - \omega_l)$, where $a_{lk}(\omega - \omega_l)$ is assumed to be sharply peaked at $\omega = \omega_l$. In this case, we can transform Eq. (24) to the time domain by expanding $\Omega_k(\omega)$ as $\Omega_k \approx \Omega_k(\omega_l) + (\omega - \omega_l)\Omega'_k(\omega_l)$, where $\Omega'_k(\omega)$ is the derivative of Ω_k with respect to the spectral parameter ω. It was found in Ref. [24] that, at least in the case of modes of a disk resonator, this derivative is not small and must be taken into account. In nonlinear terms, we simply replace $a_{lk}(\omega - \omega_l) \to \pi a_{lk}(t)\delta(\omega - \omega_l)$, which amounts to neglect of time derivatives of the nonlinear corrections. The resulting equation is

$$\begin{aligned}&\left\{-i[1 - \Omega'_k(\omega_l)]\frac{d}{dt} + [\Omega_k(\omega_l) - \omega_l]\right\}a_{lk}(t)\\ &\quad = \sum_{k'} V^{(l)}_{kk'}(t)a_{lk'}(t),\end{aligned} \qquad (28)$$

which, in the stationary limit, coincides with Eq. (27). Note that $V^{(l)}_{kk'}$ is now a slowly varying function of time via its dependence on the amplitudes $a_{l''k''}(t)$. The correction due to Ω'_k is a new term, which was not discussed in any of the previous treatments of lasing dynamics. While it does not affect the steady-state solutions, it might change their stability, and is therefore important for strongly open cavities. More detailed study of its role is outside of the scope of this article and will be presented elsewhere.

The polarization matrix $V^{(l)}_{kk'}$ in the slowly varying amplitude approximation can be presented as

$$V^{(l)}_{kk'}(t) = 2\pi\nu \int_{\mathcal{I}} d\mathbf{r}\, \epsilon^{-1}(\mathbf{r})\phi_k^*(\mathbf{r},\omega_l)\psi_{k'}(\mathbf{r},\omega_l)\eta_l(\mathbf{r},t), \qquad (29)$$

where we introduced the nonlinear susceptibility $\eta_l(\mathbf{r},t) \equiv P_l(\mathbf{r},t)/E_l(\mathbf{r},t)$ defined as the ratio of the slowly varying polarization amplitude $P_l(\mathbf{r},t)$ and the field $E_l(\mathbf{r},t)$ in the mode l. The expression for the susceptibility is found from perturbation expansion for polarization $P(\mathbf{r}, t)$, Eqs. (18) and (20), and is given by

$$\begin{aligned}\eta_l(\mathbf{r}, t) = {} & 2i\hbar\gamma_\parallel D(\omega_l)\Delta n_0(\mathbf{r}) \sum_{q \text{ odd}} A^{\frac{q+1}{2}} \sum_{l_1,\ldots,l_q}^{\mathrm{r}} \left|E_{l_2}(\mathbf{r}, t)\right|^2 \left|E_{l_4}(\mathbf{r}, t)\right|^2 \cdots \left|E_{l_{q-1}}(\mathbf{r}, t)\right|^2 D_\parallel\left(\omega_{l_1} - \omega_{l_2}\right) \\ & \times D_\parallel\left(\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \omega_{l_4}\right) \cdots D_\parallel\left(\omega_{l_1} - \omega_{l_2} + \cdots + \omega_{l_{q-2}} - \omega_{l_{q-1}}\right)\left[D\left(\omega_{l_1}\right) + D^*\left(\omega_{l_2}\right)\right]\left[D\left(\omega_{l_1} - \omega_{l_2} + \omega_{l_3}\right)\right. \\ & \left. + D^*\left(\omega_{l_2} - \omega_{l_1} + \omega_{l_4}\right)\right] \cdots \left[D(\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \cdots - \omega_{l_{q-3}} + \omega_{l_{q-2}}) + D^*\left(\omega_{l_2} - \omega_{l_1} + \omega_{l_4} - \cdots - \omega_{l_{q-4}} + \omega_{l_{q-1}}\right)\right].\end{aligned} \tag{30}$$

Here the order of nonlinearity q, introduced in Eq. (18), determines the number of different indices l_i, which take values from 1 to N_m, where N_m is the number of lasing modes. The superscript r at the sum symbol specifies that the possible values of the indices are restricted by the resonance condition

$$\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \cdots - \omega_{l_{q-1}} + \omega_{l_q} - \omega_l = 0, \tag{31}$$

which ensures cancellation of fast oscillating terms. In the absence of accidental degeneracies, this condition implies that each of the indices $l_1, l_3, \ldots, l_q$ must be equal to one of the indices $l_2, l_4, \ldots, l_{q-1}, l$. This leads, in particular, to the appearance of absolute squares of the field in the first line of Eq. (30). Moreover, the index l_q effectively drops out of the equation, since the amplitude E_{l_q} must be equal to some other amplitude E_{l_i}. It is assumed that the slowly varying field amplitudes are expressed in terms of quasimode components $a_{lk}(t)$ of the respective lth lasing mode using

$$E_l(\mathbf{r}, t) = \epsilon^{-1/2}(\mathbf{r}) \sum_k a_{lk}(t)\psi_k(\mathbf{r}, \omega_l). \tag{32}$$

By substituting Eqs. (30) and (29) into Eq. (28), one obtains a closed system of dynamic equations for a_{lk} valid to all orders in the field amplitude.

One of the fundamental difficulties of the theory of lasers is that the number N_m of lasing modes is a priori unknown and depends on the strength of the pumping and the spatial distribution of the electric field in the cavity. In Ref. [23], this value is determined by the number of possible solutions of Eq. (27) with real frequencies at a given pumping strength. However, this approach does not take into account stability of the found solutions, which can only be determined by considering the time-dependent Eq. (28). When using this equation, one could start by assuming that N_m is equal to the size of the basis of quasimodes, N_b, and study their time evolution. Those E_l that do not correspond to real lasing solutions at a given pumping would decay to zero, and the number of lasing modes would be determined a posteriori without the need for a prior knowledge of N_m. This approach is not free of difficulties either, because of possible multistable behavior and hysteresis. However, analysis of these issues is beyond the scope of this article.

V. DIAGRAMMATIC TECHNIQUE

A. Diagrammatic representation of the perturbation series

In this subsection, we present a diagrammatic technique developed to classify different nonlinear terms in Eq. (30). It should be noted that our diagrams, unlike diagrams of the field or many-particle theory, do not provide one-to-one correspondence between different terms of Eq. (30) and elements of the diagrams. The role of the diagrams here is more limited: We use them to classify different pairing possibilities for the lasing mode indices $l_1, l_2, \ldots, l_q, l$ in the perturbation series (30). Nevertheless, as it is shown, this technique allows for classification and partial summation of the classes of the terms in a manner very similar to traditional diagrammatic methods. Unlike pairing of vertices in traditional diagrammatic techniques, which reflects Wick's theorem for creation-annihilation operators or Gaussian statistics of respective random processes, the pairing procedure in the situation under consideration hinges upon the condition expressed by Eq. (31). The resonance condition guarantees the absence of the fast oscillating terms and hence the validity of the slowly changing amplitude approximation.

To construct a diagram $\widetilde{X}^0_{qj}$ of order $q = 1, 3, \ldots,$ we place $q + 1$ vertices in two columns as shown in Fig. 1. The left vertices are labeled $l_1, l_3, \ldots, l_q$, and the right vertices are labeled $l_2, l_4, \ldots, l_{q-1}, l$. The vertex l is different from the other vertices, because there is no summation over the index l in Eq. (30). After that, each vertex on the left is connected with exactly one vertex on the right. The index $j = 1, \ldots, \widetilde{N}_q$ labels all distinct connection possibilities in an arbitrary order. To obtain all diagrams of order q, we first connect the vertices by $(q + 1)/2$ horizontal links and then reshuffle the vertices, say, on the left without cutting the links. Thus, the number of

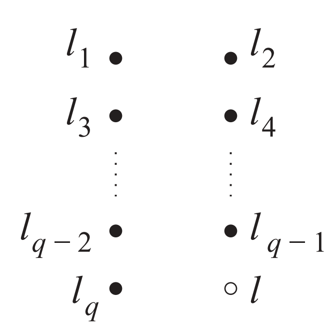

FIG. 1. Labeling of vertices in a diagram of order $q = 1, 3, 5, \ldots$.

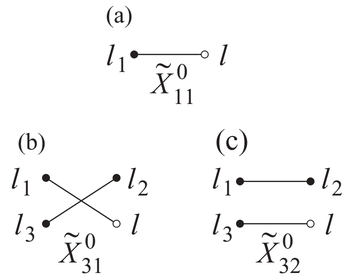

FIG. 2. First-order diagram (a) and third-order diagrams (b, c).

possible diagrams of order q is the number of permutations $\widetilde{N}_q = [(q+1)/2]!$. Diagrams for $q = 1, 3, 5$ are shown in Figs. 2 and 3.

Each diagram specifies a particular contribution to the series (30). The latter is written in the form

$$\eta_l(\mathbf{r}, t) = 2i\hbar\gamma_\parallel \Delta n_0(\mathbf{r}) D(\omega_l) \mathcal{X}_l, \tag{33}$$

$$\mathcal{X}_l \equiv \sum_{q \text{ odd}} A^{\frac{q+1}{2}} \sum_{j=1}^{\widetilde{N}_q} \widetilde{X}^0_{qj}, \tag{34}$$

where each $\widetilde{X}^0_{qj}$ represents a partial sum in $\sum^{\mathrm{r}}(\cdots)$, Eq. (30), in which pairs of indices are chosen to be equal to each other according to the links connecting respective vertices in the diagram. For example, the first three diagrams correspond to the following expressions:

$$\widetilde{X}^0_{11} = 1, \tag{35}$$

$$\widetilde{X}^0_{31} = \sum_{l_2 \neq l} \left|E_{l_2}(\mathbf{r}, t)\right|^2 D_\parallel\left(\omega_l - \omega_{l_2}\right)\left[D(\omega_l) + D^*\left(\omega_{l_2}\right)\right], \tag{36}$$

$$\widetilde{X}^0_{32} = \sum_{l_2} \left|E_{l_2}(\mathbf{r}, t)\right|^2 2\mathrm{Re}\left[D\left(\omega_{l_2}\right)\right]. \tag{37}$$

The restriction $l_2 \neq l$ in the diagram $\widetilde{X}^0_{31}$ excludes the term with $l_1 = l_2 = l_3 = l$, which enters $\widetilde{X}^0_{32}$. In general, the terms with more than two indices equal belong to the diagram in which the links connecting these indices do not cross each other. Another example includes the fifth-order terms with $l_2 = l_3 = l_4 = l_5 \neq l$, which enter $\widetilde{X}^0_{52}$ but not $\widetilde{X}^0_{51}$. The expression for arbitrary $\widetilde{X}^0_{qj}$ is given in Appendix B.

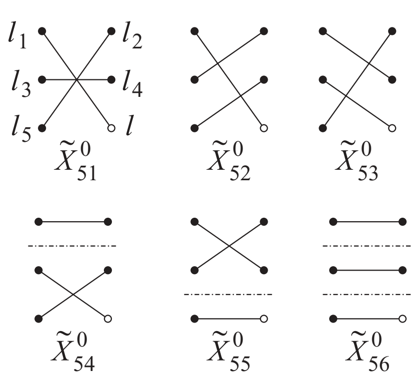

FIG. 3. Fifth-order diagrams. The dash-dotted lines are the cuts that split disconnected diagrams into connected diagrams.

B. Resummation of the diagrams

A diagram is called connected if it cannot be cut by a horizontal line without cutting a link. For instance, the diagrams $\widetilde{X}^0_{31}$, $\widetilde{X}^0_{51}$, $\widetilde{X}^0_{52}$, and $\widetilde{X}^0_{53}$ are connected, whereas the diagrams $\widetilde{X}^0_{32}$, $\widetilde{X}^0_{54}$, $\widetilde{X}^0_{55}$, and $\widetilde{X}^0_{56}$ are disconnected. To simplify the notation, we ordered all connected diagrams before the disconnected diagrams for given q. We label connected diagrams as X^0_{qj} with

$$X^0_{qj} = \widetilde{X}^0_{qj}, \quad j = 1, \ldots, N_q, \tag{38}$$

where N_q ($<\widetilde{N}_q$) is the number of connected diagrams. The horizontal cuts separate disconnected diagrams into one connected diagram containing the vertex l (denoted by an unfilled dot in the graphic representation) and several connected subdiagrams without such vertex. The latter subdiagrams are denoted as X_{qj}, $j = 1, \ldots, N_q$, where $q + 1$ is the number of vertices in the subdiagram. In place of the vertex l, these diagrams have a vertex with the index l_{q+1} that runs over all lasing modes, as the other indices l_j. For example, the diagram $\widetilde{X}^0_{55}$ consists of X^0_{11} and X_{31}, and the diagram $\widetilde{X}^0_{56}$ consists of X^0_{11} and two diagrams X_{11}, where

$$X_{11} = \sum_{l_2} \left|E_{l_2}(\mathbf{r}, t)\right|^2 2\mathrm{Re}\left[D\left(\omega_{l_2}\right)\right], \tag{39}$$

$$X_{31} = \sum_{\substack{l_2, l_4 \\ l_2 \neq l_4}} \left|E_{l_2}(\mathbf{r}, t)\right|^2 \left|E_{l_4}(\mathbf{r}, t)\right|^2 D_\parallel\left(\omega_{l_4} - \omega_{l_2}\right) \times \left[D\left(\omega_{l_4}\right) + D^*\left(\omega_{l_2}\right)\right]^2. \tag{40}$$

A general expression for X_{qj} is given in Appendix B. Note that X_{qj} is of the order $q + 1$ in the electric field. Connected diagrams contain $(q-1)/2$ nontrivial factors $D_\parallel \neq 1$.

Our diagrammatic technique possesses the basic property that disconnected diagrams are given by products of their connected parts. Thus, we can write for our examples

$$\widetilde{X}^0_{55} = X^0_{11} X_{31}, \tag{41}$$

$$\widetilde{X}^0_{56} = X^0_{11} (X_{11})^2. \tag{42}$$

The multiplicativity results from the resonance condition of the type (31) being fulfilled for each connected subdiagram (see also Appendix B). The multiplicativity property allows us to express the series (34) in terms of the connected diagrams as

$$\mathcal{X}_l = \left(\sum_{q \text{ odd}} \sum_{j=1}^{N_q} X^0_{qj}\right) \sum_{m=0}^{\infty} \left(\sum_{q \text{ odd}} \sum_{j=1}^{N_q} X_{qj}\right)^m = \frac{\sum_{q \text{ odd}} \sum_{j=1}^{N_q} X^0_{qj}}{1 - \sum_{q \text{ odd}} \sum_{j=1}^{N_q} X_{qj}}. \tag{43}$$

This resummation formula is the main result of our article.

VI. LIMITING CASES AND DISCUSSION

To make the meaning of Eq. (43) more transparent, we apply it in several well-known special cases. We start with the linear approximation in Sec. VI A and consider the effect of gain-induced coupling of passive modes [16,17]. In Sec. VI B, we reproduce semiclassical equations of the standard third-order laser theory [4,5]. In Sec. VI C, we discuss all-order nonlinear theory in the approximation of constant population inversion [15,21–23] and derive, using our theory, the first diagrammatic correction to it.

A. Linear gain-induced mode coupling

In the linear approximation to the polarization $P_l = \eta_l E_l$ [Eq. (30)], only the lowest diagram X_{11}^0 contributes to $\mathcal{X}_l$. Equation (28), where the matrix $V_{kk'}^{(l)}$ is calculated using Eqs. (29), (33), and (35), yields the following equations for the slowly varying amplitudes:

$$\sum_{k'} \left\{ \delta_{kk'} \frac{d}{dt} + i[\overline{\Omega}_{kk'}(\omega_l) - \delta_{kk'}\omega_l] \right\} a_{lk'} = 0, \tag{44}$$

where the Ω_k' term is henceforth neglected. The frequency matrix

$$\overline{\Omega}_{kk'}(\omega) = \Omega_k(\omega)\delta_{kk'} - V_{kk'}(\omega), \tag{45}$$

is modified by the linear gain term,

$$V_{kk'}(\omega) = -2i\pi\nu \frac{d^2}{\hbar\gamma_\perp} D(\omega) \int_{\mathcal{I}} d\mathbf{r} \frac{\Delta n_0(\mathbf{r})}{\epsilon(\mathbf{r})} \phi_k^*(\mathbf{r},\omega)\psi_k(\mathbf{r},\omega), \tag{46}$$

proportional to the overlap integral. Clearly, the matrices $V_{kk'}$ and $\overline{\Omega}_{kk'}$ are nondiagonal if the pump or dielectric constant are not homogeneous. In this case, the biorthogonal quasimodes of the system $\overline{\psi}_k$ and $\overline{\phi}_k$ are no longer ψ_k and ϕ_k but are determined by the right and left eigenvectors $a_{lk}^{(\mathrm{r,l})}$ of $\overline{\Omega}_{kk'}$ according to

$$\overline{\psi}_l(\mathbf{r},\omega) = \sum_{k'} a_{lk'}^{(\mathrm{r})} \psi_{k'}(\mathbf{r},\omega), \tag{47}$$

$$\overline{\phi}_l(\mathbf{r},\omega) = \sum_{k'} a_{lk'}^{(\mathrm{l})} \phi_{k'}(\mathbf{r},\omega). \tag{48}$$

The right eigenvectors $a_{lk'}^{(\mathrm{r})}$ are normal modes of Eq. (44) whose amplitudes $\overline{a}_l(t)$ obey the equation

$$\dot{\overline{a}}_l + i[\overline{\Omega}_l(\omega_l) - \omega_l]\overline{a}_l = 0, \tag{49}$$

where $\overline{\Omega}_l(\omega)$ are eigenvalues of $\overline{\Omega}_{kk'}(\omega)$. We recall that electric field in the mode l has a time dependence $\overline{a}_l(t)\exp(-i\omega_l t)$. Thus, the lasing frequency ω_l is determined from the requirement

$$\mathrm{Re}[\overline{\Omega}_l(\omega_l)] = \omega_l, \tag{50}$$

and the threshold condition for this mode is

$$\mathrm{Im}[\overline{\Omega}_l(\omega_l)] = 0. \tag{51}$$

As follows from Eq. (49), the mode amplitudes diverge exponentially above the threshold. Hence, applicability of the linear approximation is limited to the pump strength below or at the threshold. However, the basis of normal modes can be used as a starting point in nonlinear theories.

B. Third-order theory

To obtain an approximation to $\mathcal{X}_l$ of the third order in the field, we keep the diagrams X_{11}^0 and X_{31}^0 in the numerator and the diagram X_{11} in the denominator of Eq. (43) and expand the latter:

$$\mathcal{X}_l \approx X_{11}^0 + X_{11}^0 X_{11} + X_{31}^0. \tag{52}$$

It is convenient to write lasing equations in the basis of quasimodes $\overline{\psi}_k$ and $\overline{\phi}_k$ that diagonalize the linear part. Because of nonlinear effects, the lasing modes above the threshold,

$$E_l(\mathbf{r},t) = \epsilon^{-1/2}(\mathbf{r}) \sum_k \overline{a}_{lk}(t)\overline{\psi}_k(\mathbf{r},\omega_l), \tag{53}$$

are in general linear combinations of individual quasimodes. The equation for the amplitudes $\overline{a}_{lk}(t)$ follows from Eq. (28), after taking into account the results of linear theory (Sec. VI A), and has the form

$$\begin{aligned}
&\dot{\overline{a}}_{lk} + i[\overline{\Omega}_k(\omega_l) - \omega_l]\overline{a}_{lk} \\
&\quad = -\frac{\pi\nu}{\hbar\gamma_\parallel}\left(\frac{d^2}{\hbar\gamma_\perp}\right)^2 D(\omega_l) \int_{\mathcal{I}} d\mathbf{r} \frac{\Delta n_0(\mathbf{r})}{\sqrt{\epsilon(\mathbf{r})}} \overline{\phi}_k^*(\mathbf{r},\omega_l) E_l(\mathbf{r},t) \\
&\qquad \times \sum_{l'=1}^{N_{\mathrm{m}}} |E_{l'}(\mathbf{r},t)|^2 \{2\mathrm{Re}[D(\omega_{l'})] + (1-\delta_{ll'})D_\parallel(\omega_l - \omega_{l'}) \\
&\qquad \times [D(\omega_l) + D^*(\omega_{l'})]\}, \quad k = 1,\ldots,N_{\mathrm{b}},
\end{aligned} \tag{54}$$

where N_{b} is the size of the basis of quasimodes and N_{m} is the number of lasing modes. The lasing frequencies ω_l and the mode thresholds need to be determined from these $N_{\mathrm{b}} \times N_{\mathrm{m}}$ equations in the stationary regime $\dot{\overline{a}}_{lk} = 0$ using, for example, a self-consistent procedure described in Ref. [21].

In some cases, the standard assumption of a traditional lasing theory that the lasing modes coincide with the quasimodes of the cavity remain valid. In this case, the total number of Eqs. (54) is reduced to N_{b} because the amplitudes are approximated as $\overline{a}_{lk}(t) = \overline{a}_l(t)\delta_{lk}$. By representing $\overline{a}_l(t) = \sqrt{I_l}\exp(i\overline{\varphi}_l)$ and separating the real and imaginary parts in Eq. (54), we obtain $2N_{\mathrm{b}}$ real equations for the intensities I_l and phases $\overline{\varphi}_l$,

$$\begin{aligned}
&\dot{I}_l - 2\mathrm{Im}[\overline{\Omega}_l(\omega_l)]I_l \\
&\quad = -\frac{2\pi\nu}{\hbar\gamma_\parallel}\left(\frac{d^2}{\hbar\gamma_\perp}\right)^2 I_l \mathrm{Re}\left[D(\omega_l)\sum_{l'} B_{ll'} I_{l'}\{\cdots\}\right],
\end{aligned} \tag{55}$$

$$\begin{aligned}
&\dot{\overline{\varphi}}_l + \mathrm{Re}[\overline{\Omega}_l(\omega_l)] - \omega_l \\
&\quad = -\frac{\pi\nu}{\hbar\gamma_\parallel}\left(\frac{d^2}{\hbar\gamma_\perp}\right)^2 \mathrm{Im}\left[D(\omega_l)\sum_{l'} B_{ll'} I_{l'}\{\cdots\}\right],
\end{aligned} \tag{56}$$

The terms enclosed in the braces are the same as those in Eq. (54). We defined overlap integrals for the quasimodes as

$$B_{ll'} = \int_{\mathcal{I}} d\mathbf{r} \frac{\Delta n_0(\mathbf{r})}{[\epsilon(\mathbf{r})]^2} \overline{\phi}_l^*(\mathbf{r},\omega_l)\overline{\psi}_l(\mathbf{r},\omega_l)|\overline{\psi}_{l'}|^2. \tag{57}$$

The lasing frequencies ω_l are determined, together with the stationary intensities, from Eqs. (55) and (56) in the stationary regime $\dot{I}_l = 0$ and $\dot{\varphi}_l = 0$. The rate Eqs. (55) and frequency Eqs. (56) generalize the standard third-order semiclassical theory with the self- and cross-saturation terms [4,5] to systems with strong openness and arbitrary distribution of refractive index.

C. Constant-inversion approximation and corrections

In many physical situations, the population inversion $\Delta n(\mathbf{r}, t)$ is approximately constant in time. Oscillations of the population (population pulsations) are responsible for the terms proportional to $D_\parallel(\omega_l - \omega_{l'})$, $l \neq l'$, in the expansion of the nonlinear susceptibility (30) and, hence, in the expressions for the diagrams. By comparing the terms in the braces in Eq. (54), we can conclude that the population pulsations can be neglected if $D_\parallel(\omega_l - \omega_{l'}) \ll 1$, that is, $|\omega_l - \omega_{l'}| \gg \gamma_\parallel$ for all lasing frequencies $\omega_l \neq \omega_{l'}$.

In the approximation of constant inversion, only the diagrams X_{11}^0 and X_{11}, which do not contain the $D_\parallel$ functions, contribute to $\mathcal{X}_l$ (43). Equation (28) for the coefficients of expansion (32) of the electric field becomes

$$\dot{a}_{lk} + i[\Omega_k(\omega_l) - \omega_l]a_{lk} = 2\pi\nu \frac{d^2}{\hbar\gamma_\perp} \int_{\mathcal{I}} d\mathbf{r} \frac{\Delta n_0(\mathbf{r})}{\sqrt{\epsilon(\mathbf{r})}} \times \frac{\phi_k^*(\mathbf{r}, \omega_l) E_l(\mathbf{r}, t)}{1 + [d^2/(2\hbar^2\gamma_\perp\gamma_\parallel)] \sum_{l'} |E_{l'}(\mathbf{r}, t)|^2 2\mathrm{Re}[D(\omega_{l'})]}. \quad (58)$$

In contrast to Eq. (54), the linear contribution here is not diagonalized and is contained in the right-hand side.

Equation (58) is valid in all orders in nonlinearity if the population inversion is constant. With the help of Eq. (43), it is straightforward to write out the corrections resulting from the population pulsations. The terms of the first order in $D_\parallel \ll 1$ are contained in the diagrams X_{31}^0 and X_{31} [Eqs. (36) and (40)], so that $\mathcal{X}_l$ can be approximated as

$$\mathcal{X}_l \approx \frac{X_{11}^0 + X_{31}^0}{1 - X_{11} - X_{31}}. \quad (59)$$

These corrections modify both the numerator and denominator of Eq. (58), which can now be presented as

$$\dot{a}_{lk} + i[\Omega_k(\omega_l) - \omega_l]a_{lk} = 2\pi\nu \frac{d^2}{\hbar\gamma_\perp} \int_{\mathcal{I}} d\mathbf{r} \frac{\Delta n_0(\mathbf{r})}{\sqrt{\epsilon(\mathbf{r})}} \frac{\phi_k^*(\mathbf{r}, \omega_l) E_l(\mathbf{r}, t)(1 - \Upsilon_\mathrm{n})}{1 + [d^2/(2\hbar^2\gamma_\perp\gamma_\parallel)] \sum_{l'} |E_{l'}(\mathbf{r}, t)|^2 2\mathrm{Re}[D(\omega_{l'})](1 - \Upsilon_\mathrm{d})}, \quad (60)$$

where

$$\Upsilon_\mathrm{n} = \frac{d^2}{2\hbar^2\gamma_\perp\gamma_\parallel} \sum_{l' \neq l} |E_{l'}(\mathbf{r}, t)|^2 D_\parallel(\omega_l - \omega_{l'})[D(\omega_l) + D^*(\omega_{l'})], \quad (61)$$

$$\Upsilon_\mathrm{d} = \frac{d^2}{2\hbar^2\gamma_\perp\gamma_\parallel} \sum_{l'' \neq l'} |E_{l''}(\mathbf{r}, t)|^2 D_\parallel(\omega_{l''} - \omega_{l'}) \times \frac{[D(\omega_{l''}) + D^*(\omega_{l'})]^2}{2\mathrm{Re}[D(\omega_{l'})]}. \quad (62)$$

Taking into account that the electric field in the lasing modes has a typical magnitude of $\hbar\sqrt{\gamma_\perp\gamma_\parallel}/d$, one can see that deviations from the constant-inversion approximation are of the order of $D_\parallel(\omega_l - \omega_{l'})$ [the term $D(\omega_l)$ is of the order of unity because frequencies of lasing modes are concentrated within the width of the gain profile]. The difference between lasing frequencies can be estimated as $|\omega_l - \omega_{l'}| \approx \gamma_\perp/N_\mathrm{m}$. Then the condition $D_\parallel \ll 1$ can be expressed as $N_\mathrm{m}\gamma_\parallel/\gamma_\perp \ll 1$. Given that $\gamma_\parallel$ is usually several orders of magnitude smaller than $\gamma_\perp$, this condition is in most situations fulfilled. It was reported in Ref. [22], however, that nonlinear interaction between lasing modes can push their frequencies toward each other, making the intermode spectral interval much smaller than the typical value given previously. Such pairs of modes can result in significant corrections to the constant-population approximation. Adding to the expansion additional connected diagrams with up to $q + 1$ vertices, one can improve the constant-population approximation by constructing lasing equations valid in the order $(q - 1)/2$ in $D_\parallel$.

VII. CONCLUSIONS

We presented a diagrammatic semiclassical laser theory valid in all orders of electric field. The original perturbation series in the powers of the field can be resummed in terms of a certain class of diagrams, the connected diagrams. The resummation allows one to construct a controlled expansion in the small parameter $\gamma_\parallel/\gamma_\perp$, which is a measure of population pulsations, while treating the nonlinearity exactly. Our lasing equations generalize the all-order nonlinear equations in the constant-inversion approximation and the third-order equations with population-pulsation terms. The use of CF quasimodes as basis functions makes it possible to apply the theory to strongly open and irregular systems, such as random lasers and lasers with chaotic resonators.

ACKNOWLEDGMENTS

Financial support was provided by the Deutsche Forschungsgemeinschaft via Sonderforschungsbereich/Transregio 12 and Forschergruppe 557 grants (OZ) and by Professional Staff Congress–City University of New York via Grants No. 62680-00 39 and No. 61788-00 39 (LD).

APPENDIX A: DERIVATION OF EQS. (20) AND (21)

For a few low values of q, the validity of Eqs. (20) and (21) can be checked directly. To prove these relations by induction, we assume that $\Delta n_\omega^{(q-1)}$ is given by Eq. (21), then derive $P_\omega^{(q)}$ (20), and, finally, obtain $\Delta n_\omega^{(q+1)}$.

According to Eq. (15),

$$P_\omega^{(q)} = i\hbar\gamma_\parallel \frac{A}{\pi} D(\omega) \int d\omega' E_{\omega'} \Delta n_{\omega-\omega'}^{(q-1)}. \tag{A1}$$

By substituting $\Delta n_\omega^{(q-1)}$ from Eq. (21), we immediately see that the factor before the integral in Eq. (20) is reproduced. To calculate the integral, we consider separately the two contributions: $\Delta n_{\omega-\omega'}^{(q-1)} = \widetilde{\Delta n}_{\omega-\omega'}^{(q-1)} + [\widetilde{\Delta n}_{\omega'-\omega}^{(q-1)}]^*$, where $\widetilde{\Delta n}_\omega^{(q-1)}$ is given explicitly by Eq. (21) and $[\widetilde{\Delta n}_{-\omega}^{(q-1)}]^*$ is c.c. $(\omega \to -\omega)$. When integrating $\widetilde{\Delta n}_{\omega-\omega'}^{(q-1)}$ we introduce a new variable,

$$\omega_{q-1} = \omega' - \omega + \omega_1 - \omega_2 + \cdots - \omega_{q-3} + \omega_{q-2}. \tag{A2}$$

Then the integral over ω' becomes

$$\begin{aligned} &\int d\omega' D_\parallel(\omega-\omega') E_{\omega'} E^*_{\omega'-\omega+\omega_1-\omega_2+\cdots-\omega_{q-3}+\omega_{q-2}} \\ &= \int d\omega_{q-1} D_\parallel(\omega_1 - \omega_2 + \cdots + \omega_{q-2} - \omega_{q-1}) \\ &\quad \times E_{\omega-\omega_1+\omega_2-\cdots-\omega_{q-2}+\omega_{q-1}} E^*_{\omega_{q-1}}. \end{aligned} \tag{A3}$$

A comparison with Eq. (20) shows that the $\widetilde{\Delta n}_{\omega-\omega'}^{(q-1)}$ contribution yields the part of the $P_\omega^{(q)}$ integrand proportional to $D(\omega_1 - \omega_2 + \cdots + \omega_{q-2})$. When integrating $[\widetilde{\Delta n}_{\omega'-\omega}^{(q-1)}]^*$, we first exchange the labels $\omega_1 \leftrightarrow \omega_2$, $\omega_3 \leftrightarrow \omega_4, \ldots, \omega_{q-2} \to \omega_{q-1}$ and then define the variable

$$\omega_{q-2} = \omega - \omega' - \omega_1 + \omega_2 - \cdots + \omega_{q-3} + \omega_{q-1}. \tag{A4}$$

By transforming the integral over ω' as

$$\begin{aligned} &\int d\omega' D_\parallel(\omega-\omega') E_{\omega'} E_{\omega-\omega'-\omega_1+\omega_2-\cdots+\omega_{q-3}+\omega_{q-1}} \\ &= \int d\omega_{q-2} D_\parallel(\omega_1 - \omega_2 + \cdots + \omega_{q-2} - \omega_{q-1}) \\ &\quad \times E_{\omega-\omega_1+\omega_2-\cdots-\omega_{q-2}+\omega_{q-1}} E_{\omega_{q-2}}, \end{aligned} \tag{A5}$$

we obtain the part of the $P_\omega^{(q)}$ integrand proportional to $D^*(\omega_2 - \omega_1 + \cdots + \omega_{q-1})$.

Next, we derive $\Delta n_\omega^{(q+1)}$ using Eq. (16),

$$\Delta n_\omega^{(q+1)} = \frac{-i}{\pi\hbar\gamma_\parallel} D_\parallel(\omega) \int d\omega' E^*_{\omega'-\omega} P_{\omega'}^{(q)} + \text{c.c.}(\omega \to -\omega). \tag{A6}$$

Clearly, the factor before the integral in Eq. (21) follows after substitution of $P_{\omega'}^{(q)}$ (20). By introducing the new variable,

$$\omega_q = \omega' - \omega_1 + \omega_2 - \cdots - \omega_{q-2} + \omega_{q-1}, \tag{A7}$$

we rewrite the ω' integral

$$\begin{aligned} &\int d\omega' D(\omega') E^*_{\omega'-\omega} E_{\omega'-\omega_1+\omega_2-\cdots-\omega_{q-2}+\omega_{q-1}} \\ &= \int d\omega_q D(\omega_1 - \omega_2 + \cdots - \omega_{q-1} + \omega_q) \\ &\quad \times E^*_{-\omega+\omega_1-\omega_2+\cdots-\omega_{q-1}+\omega_q} E_{\omega_q}. \end{aligned} \tag{A8}$$

This completes the proof of Eqs. (20) and (21).

APPENDIX B: GENERAL EXPRESSIONS FOR DIAGRAMS

The diagrams $\widetilde{X}_{qj}^0$ $(q = 3, 5, \ldots)$ are given by the following analytical expression:

$$\begin{aligned} \widetilde{X}_{qj}^0 = \sum_{\widetilde{X}_{qj}^0} &|E_{l_2}(\mathbf{r},t)|^2 |E_{l_4}(\mathbf{r},t)|^2 \cdots |E_{l_{q-1}}(\mathbf{r},t)|^2 D_\parallel(\omega_{l_1} - \omega_{l_2}) D_\parallel(\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \omega_{l_4}) \cdots D_\parallel(\omega_{l_1} - \omega_{l_2} + \cdots + \omega_{l_{q-2}} - \omega_{l_{q-1}}) \\ &\times [D(\omega_{l_1}) + D^*(\omega_{l_2})][D(\omega_{l_1} - \omega_{l_2} + \omega_{l_3}) + D^*(\omega_{l_2} - \omega_{l_1} + \omega_{l_4})] \cdots [D(\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \cdots - \omega_{l_{q-3}} + \omega_{l_{q-2}}) \\ &+ D^*(\omega_{l_2} - \omega_{l_1} + \omega_{l_4} - \cdots - \omega_{l_{q-4}} + \omega_{l_{q-1}})]. \end{aligned} \tag{B1}$$

The symbol $\sum_{\widetilde{X}_{qj}^0}$ denotes a summation over the lasing-mode indices $l_2, l_4, \ldots, l_{q-1} = 1, \ldots, N_\text{m}$ according to these rules: (i) if the indices l_i and l_j are connected in the diagram $\widetilde{X}_{qj}^0$, set $l_i = l_j$, and (ii) the terms with four or more indices $l_1, l_2, \ldots, l_q, l$ equal are excluded unless the links connecting the affected vertices do not intersect.

We denote by X_{qj}^0 connected diagrams and subdiagrams containing the vertex l. With the ordering of $\widetilde{X}_{qj}^0$ such that the connected diagrams come before the disconnected diagrams for a given q, we can identify $X_{qj}^0 = \widetilde{X}_{qj}^0$ $(j \leqslant N_q)$, where N_q is the number of connected diagrams. The subdiagrams X_{qj} without the special vertex have a variable index l_{q+1} in place of the fixed index l. The analytical expression for these diagrams has the form

$$\begin{aligned} X_{qj} = \sum_{X_{qj}} &|E_{l_2}(\mathbf{r},t)|^2 |E_{l_4}(\mathbf{r},t)|^2 \cdots |E_{l_{q+1}}(\mathbf{r},t)|^2 D_\parallel(\omega_{l_1} - \omega_{l_2}) D_\parallel(\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \omega_{l_4}) \cdots D_\parallel(\omega_{l_1} - \omega_{l_2} + \cdots + \omega_{l_{q-2}} - \omega_{l_{q-1}}) \\ &\times [D(\omega_{l_1}) + D^*(\omega_{l_2})][D(\omega_{l_1} - \omega_{l_2} + \omega_{l_3}) + D^*(\omega_{l_2} - \omega_{l_1} + \omega_{l_4})] \cdots [D(\omega_{l_1} - \omega_{l_2} + \omega_{l_3} - \cdots - \omega_{l_{q-1}} + \omega_{l_q}) \\ &+ D^*(\omega_{l_2} - \omega_{l_1} + \omega_{l_4} - \cdots - \omega_{l_{q-2}} + \omega_{l_{q+1}})]. \end{aligned} \tag{B2}$$

The sum $\sum_{X_{qj}}$ is defined analogously to the previous sum $\sum_{\widetilde{X}^0_{qj}}$. Note that the diagrams $\widetilde{X}^0_{qj}$ and X^0_{qj} are of the order $q-1$ in the electric field, whereas the subdiagrams X_{qj} are of the order $q+1$.

To show the multiplicativity property, let us assume that a disconnected diagram $\widetilde{X}^0_{qj}$ can be cut in two possibly disconnected subdiagrams, $\widetilde{X}^0_{q'j'}$ and $X_{q''j''}$, such that $q = q' + q'' + 1$. We label the vertices of $X_{q''j''}$ as $l_1, \ldots, l_{q''+1}$ and identify them with the first $q''+1$ vertices of the diagram $\widetilde{X}^0_{qj}$. The last $q'+1$ vertices of $\widetilde{X}^0_{qj}$, $l_{q''+2}, \ldots, l_q, l$, are also the vertices of $X^0_{q'j'}$. We need to show that, in the sum $\sum_{\widetilde{X}^0_{qj}}$, the two groups of indices can be split between the sums $\sum_{X_{q''j''}}$ and $\sum_{X^0_{q'j'}}$, respectively. This would mean that the arguments of the functions D and $D_\parallel$ can contain only the indices belonging to one of the groups. This is indeed the case, because the arguments having less than $q''+1$ frequencies contain only the indices from the first group, whereas in the arguments with more frequencies the first $q''+1$ frequencies cancel because of the cut as

$$\omega_1 + \omega_3 + \cdots + \omega_{q''} = \omega_2 + \omega_4 + \cdots + \omega_{q''+1}. \tag{B3}$$

According to this equality, the number of nontrivial factors $D_\parallel \neq 1$ in Eqs. (B1) and (B2) is $(q-1)/2$ minus the number of cuts (in a disconnected diagram).

[1] S. V. Frolov, Z. V. Vardeny, A. Zakhidov, and R. H. Baughman, Opt. Commun. **162**, 241 (1999).

[2] H. Cao, Y. G. Zhao, S. T. Ho, E. W. Seelig, Q. H. Wang, and R. P. H. Chang, Phys. Rev. Lett. **82**, 2278 (1999).

[3] W. Fang, H. Cao, and G. S. Solomon, Appl. Phys. Lett. **90**, 081108 (2007).

[4] M. Sargent III, M. O. Scully, and W. E. Lamb Jr., *Laser Physics* (Addison-Wesley, Reading, MA, 1974).

[5] H. Haken, *Laser Theory* (Springer-Verlag, Berlin, 1984).

[6] A. Fox and T. Li, IEEE J. Quantum Electron. **4**, 460 (1968).

[7] A. E. Siegman, Phys. Rev. A **39**, 1253 (1989).

[8] A. E. Siegman, Phys. Rev. A **39**, 1264 (1989).

[9] N. Moiseyev, Phys. Rep. **302**, 212 (1998).

[10] E. S. C. Ching, P. T. Leung, A. Maassen van den Brink, W. M. Suen, S. S. Tong, and K. Young, Rev. Mod. Phys. **70**, 1545 (1998).

[11] S. M. Dutra and G. Nienhuis, Phys. Rev. A **62**, 063805 (2000).

[12] C. Viviescas and G. Hackenbroich, Phys. Rev. A **67**, 013805 (2003).

[13] C. Viviescas and G. Hackenbroich, J. Opt. B: Quantum Semiclass. Opt. **6**, 211 (2004).

[14] P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), Vol. 1.

[15] H. E. Türeci, A. D. Stone, and B. Collier, Phys. Rev. A **74**, 043822 (2006).

[16] L. I. Deych, Phys. Rev. Lett. **95**, 043902 (2005).

[17] L. Deych, in *Complex Mediums VI: Light and Complexity*, edited by M. W. McCall, G. Dewar, and M. A. Noginov (SPIE, Bellingham, WA, 2005), Vol. 5924, p. 59240B.

[18] T. S. Misirpashaev and C. W. J. Beenakker, Phys. Rev. A **57**, 2041 (1998).

[19] G. Hackenbroich, J. Phys. A **38**, 10537 (2005).

[20] O. Zaitsev, L. Deych, and V. Shuvayev, Phys. Rev. Lett. **102**, 043906 (2009).

[21] H. E. Türeci, L. Ge, S. Rotter, and A. D. Stone, Science **320**, 643 (2008). See also the supporting online material.

[22] H. E. Türeci, A. D. Stone, L. Ge, S. Rotter, and R. J. Tandy, Nonlinearity **22**, C1 (2009).

[23] H. E. Türeci, A. D. Stone, and L. Ge, Phys. Rev. A **76**, 013813 (2007).

[24] V. Shuvayev and L. Deych (private communication).